

Probing quantum nanostructures with near-field optical microscopy and (vice versa)

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Abstract

A theory is presented to show how near-field optical microscopy can be used to probe quantum nanostructures. Calculations are done for a quantum dot. Results for different tip/dot configurations and sizes show that near-field excitation can enhance light-hole transitions, excite selection-rule breaking transitions with rates comparable to allowed transitions, and map electron-hole pair wave functions. Conversely, dot response can be used to characterize tip near-fields.

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Traditional far-field optical spectroscopy has been used extensively to probe quantum nanostructures. Wavelength and polarization are varied to access different transitions. However, spatial resolution, diffraction limited to $\lambda/2$, is much larger than most nanostructures and only dipole allowed transitions can be excited. Recently, near-field scanning optical microscopy (NSOM) has been studied intensively to achieve spatial resolution much better than $\lambda/2$. In NSOM, an optical fiber that has been tapered to a 10-100 nm wide tip and metal coated with an aperture at the tip is used as a localized light source or collector. NSOM has been exploited to investigate single molecules [1,2], T-shaped quantum wires [3], and 10-nm wide self-assembled CdSe quantum dots [4]. In these cases the nanosystems were much smaller than the NSOM probe field. NSOM spatial resolution was exploited only to localize the excitation on a small collection of structures. Development of theory for NSOM excitation of very small systems has begun [5,6].

Excitons confined at quantum well width fluctuations with a lateral size *comparable* to the probe field spread have also been studied. NSOM was used to isolate single excitons in these systems, but spatial resolution was degraded by lateral exciton diffusion [7]. Isolated excitons were also studied by collecting photoemission that escaped through small holes in a metal layer on the surface of the quantum well sample [8,9]. In this case, the spatial configuration of the aperture and confined exciton was fixed. No spatial information about confined systems was obtained either in these cases.

In this letter, a simple theory is presented to show how NSOM can be used to probe nanostructures with sizes similar to the near-field spread. As an example, a dot scanned by an NSOM tip is considered. With NSOM, wavelength, polarization, *and* tip/sample separation can be used to control the transitions that are studied. The tip/sample separation can be varied to modify transition rates and access transitions that are forbidden in far-field spectroscopy. These effects depend on the probe-field spread being comparable to the nanostructure size. I will show that typical NSOM probes are sufficiently localized to produce observable effects for confined systems.

A schematic of the experiment to be modeled is shown in Fig. 1. Simple but realistic

models for the tip field, the dot, and the dot confined states are used to reduce the complexity of the calculations and to focus on the essentials of this experiment rather than the details of a particular system. The tip field incident on the sample is described by the Bethe-Bouwkamp model [10,11]. In this approximation, the tip near-field is modeled by the near-field transmitted by a circular aperture, radius a , in a perfectly conducting metal screen due to a plane-wave field incident normal to the screen. This model has been used successfully to understand NSOM measurements [1,12]. Implicit in this model is the assumption that local fields due to the sample do not drastically alter the incident tip field. The importance of local fields depends on the dielectric contrast of the sample. The dot must be close to the top of the sample so that tip evanescent fields can couple to the dot. Here, I assume that there is little dielectric contrast between the dot and its substrate. In that case, the substrate mainly refracts the tip field.

Optical transition rates are determined by Fermi's golden rule using the spatially varying tip field to excite transitions. The dot/probe coupling depends on the overlap of the dot states with the probe field and on the atomic transition matrix elements between confined electron and hole states. The dot is taken to be rectangular, with lateral widths L_x and L_y much greater than its thickness L_z . Infinite barriers are used to model the confinement. Using finite barriers instead would just increase the effective size of the dot states. Confined electron and hole envelop functions are found by use of single-band effective mass theory. The electron conduction states (e) are constructed from s atomic states. For the atomic state of the confined hole, I use the $J = 3/2$ atomic states obtained with a 4-band Luttinger model for holes in a (001) quantum well with $k_x = k_y = 0$. For heavy holes (hh), the atomic state is $\mp\sqrt{1/2}(|x\rangle \pm i|y\rangle)|\pm 1/2\rangle_s$; for light holes (lh), $\sqrt{2/3}|z\rangle|\pm 1/2\rangle_s + \sqrt{1/6}(|x\rangle \pm i|y\rangle)|\mp 1/2\rangle_s$. I assume that the atomic state is unaffected by lateral confinement, which is reasonable for $L_x, L_y \gg L_z$. The hh and lh response will be different, because the lh contains a $|z\rangle$ component and can couple to electric fields in the z -direction, E_z . E_z is not present when probing with a plane wave at normal incidence from the far-field, but is a significant component of the tip near-field. The optical excitations are electron-hole pair states formed

from the confined single-particle states. In this paper I ignore pair interaction. These effects can be important in wide dots and will be included in a later study.

To begin, it is useful to remember how a dot responds to a linearly polarized plane wave incident normal to the dot from the far-field. Lateral quantum numbers are conserved ($n_{ex} = n_{hx}$ and $n_{ey} = n_{hy}$), because the incident field has no lateral variation. The z quantum number n_z is conserved ($n_{ez} = n_{hx}$), because $\lambda \gg L_z$. Also, the hh transition rate is 3 times the lh rate because there is no E_z to couple to the lh.

To understand how NSOM can be used to probe nanostructures, one must know the tip near-fields. In the Bethe-Bouwkamp model [1,10,11], a linearly polarized plane wave incident on an aperture drives charge around the aperture, producing an oscillating dipole moment in the direction of the incident polarization. The near field can be found in the quasistatic limit and is the field of the instantaneous dipole. Near the aperture center and near the aperture plane, the near field is in the direction of the incident polarization (see Fig. 1). Away from the center there is a weak field in the direction transverse to polarization and to z . Near the aperture edges, the dipole field is dominantly along z . Thus the NSOM field has two large components; one along the incident polarization, which maintains its symmetry across the aperture but drops off rapidly at the edges, and E_z , which is large at the edges, small in the center, and changes sign going across the aperture. Transitions that couple to E_x and E_y will be strongly excited when the tip is over the dot. Transitions excited by E_z will be stronger when the tip edge is over the dot center. Transitions that normally are forbidden because they do not conserve parity along the polarization can be excited by E_z . Other symmetry-forbidden transitions become allowed when the tip is not centered over the dot. The coupling to E_z and the forbidden transitions are more important for smaller apertures.

To show that these effects can be significant, I present four examples. The field E_i incident on the aperture is x polarized with wavelength $\lambda = 822$ nm, corresponding to transitions near 1.5eV. The dot has $L_x = L_y = 100$ nm and $L_z = 10$ nm. Figures 2-5 present rates for different transitions as a function of the tip/dot separation, defined in Fig. 1, as the tip is scanned at a height z_t above the dot center line in the indicated direction.

Figure 2 shows rates for transitions that conserve quantum number with $n_x = n_z = 1$. When the tip is over the dot center ($x = 0$), hh rates are 3 times the corresponding lh rates, as in the far-field, because there is no coupling to E_z when dot and tip coincide. As the tip moves to the dot edge ($x = 50$ nm), hh rates follow E_x and decrease rapidly. The lh rates respond to E_z as well as E_x and increase slowly until the tip edge passes beyond the dot center. When the tip is outside the dot, lh transitions are much stronger than hh transitions, reversing the usual ordering of these rates. This reversal occurs where lh rates are still significant and should be observable. When the tip scans parallel to the incident polarization and probes states that vary monotonically (no nodes) away from the dot center in this direction, the variations in the rates map the near field and characterize tip size. In this case, the position of peak lh rate indicates the tip radius.

Oscillations in pair-state wavefunctions can be probed by scanning the tip along the oscillations. Rates for transitions with $n_x \geq 1$ are shown in Fig. 3 for the configuration used in Fig. 2, scanning along the polarization, but with $z_t = 2.5$ nm to increase coupling to the near field. Oscillations in the rates are due to variations in the near field along the polarization direction and oscillations in the pair wave functions in this direction. To separate these two effects, one can scan perpendicular to the polarization, in the y direction, where there are no oscillations in E_x or E_z . Fig. 4 shows a scan along y for transitions with $n_x = 1$ but $n_y \geq 1$. As in Fig. 2, $z_t = 10$ nm, but now $a = 12.5$ nm so the spread of the near field is smaller than the dot and the tip acts more like a point source. Oscillations in the rates are now due only to oscillations in the pair wave functions. These oscillations are comparable to the magnitudes of the rates and should be observable. Oscillations are further enhanced by reducing z_t .

Scans done in different directions, for different polarizations, and for different z_t control how transitions are probed. Reducing z_t increases the near field and the coupling to dot states when the tip is over the dot. Increasing z_t spreads out the tip field and enhances coupling to transitions which are important when the tip is outside the dot.

Transitions that do not conserve lateral quantum number and would be weak in far-field

spectroscopy can be excited by the near field because the lateral spatial variation of the tip near field is comparable to the dot size. Transitions between confined states with different lateral quantum numbers can be strongly excited by a field with a lateral variation only if the lateral wavevector of the field is so large that the field is evanescent along z [13]. Thus one must use near fields to excite these transitions. Figure 5 shows rates for transitions between the electron ground state and hole states with either $n_{hx} > 1$ or $n_{hy} > 1$. The same configuration is used as in Fig. 2. Normally forbidden transitions to $n_{hx} = 2$ hh and lh states are strongly enhanced when the tip is moved from the dot center to the peak in the hole wave function. Other scan directions and configurations enhance other far-field forbidden transitions. These transitions are weaker than number conserving transitions (compare with Fig. 2). However, near the dot edge they are no more than three times weaker and should be observable. Reducing z_t increases the rates for far-field forbidden transitions.

In summary, a theory has been presented to show how near-field optical microscopy can be used to probe nanostructures. Scanning a near-field across a nanostructure of similar size can selectively enhance light hole transitions, selectively excite far-field forbidden transitions with rates comparable to allowed transitions, and map electron-hole pair wave functions. Conversely, the dot response can be used to map tip near-fields.

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FIGURES

FIG. 1. Schematic of a quantum dot scanned by an NSOM tip. Simplifications in the theory are indicated. The aperture near field is shown.

FIG. 2. Transition rates when an NSOM tip (radius $a = 50$ nm) is scanned along x at a height $z_t = 10$ nm above the dot center line. Quantum-number-conserving transitions with $n_y \geq 1$ are shown.

FIG. 3. Transition rates when an NSOM tip is scanned along x above the dot center line: $a = 50$ nm, $z_t = 2.5$ nm. Quantum-number-conserving transitions with $n_x \geq 1$ are shown.

FIG. 4. Transition rates when an NSOM tip is scanned along y above the dot center line: $a = 12.5$ nm, $z_t = 10$ nm. Quantum-number-conserving transitions with $n_y \geq 1$ are shown.

FIG. 5. Transition rates when an NSOM tip is scanned along x above the dot center line: $a = 50$ nm, $z_t = 10$ nm. Transitions between electron ground state and hole excited states are shown.









